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FINAL DESIGN FOR THERMAL EPITHERMAL EXPERIMENTS (TEX) WITH ZPPR PLUTONIUM/ALUMINUM PLATES WITH POLYETHYLENE AND TANTALUM

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ABSTRACT

Lawrence Livermore National Laboratory has completed the final design for the first phase of the Thermal Epithermal eXperiments (TEX), which focused on critical configurations composed of plutonium/aluminum Zero Power Physics Reactor (ZPPR) plates moderated by polyethylene with and without a tantalum diluent. ^{239}Pu and ^{240}Pu were identified as being the number one and two nuclear data need by the nuclear criticality safety and nuclear data communities, with special emphasis placed on performance issues of the Pu cross sections in the intermediate energy region. Preliminary TEX design recommended tantalum as the first diluent to pursue for final design, as it represented a high priority data need, showed the highest sensitivity and largest contribution to uncertainty with the Pu ZPPR system, and a large number of high quality Ta plates from the ZPPR inventory in Idaho was readily available.

The goals of the new critical experiments are 1) to create assemblies that span a wide range of fission energy spectra, from thermal (below 0.625 eV), through the intermediate (0.65 eV to 100 keV), and to fast energies (above 100 keV), and 2) to easily modify the assemblies to include high priority materials like tantalum. A total of ten critical configurations were designed as part of CED-2. Five TEX experiments (Experiments 1-5) were designed to establish baseline configurations with the Pu ZPPR plates covering the thermal, intermediate, and fast fission energy regimes. For all five experiments, the ZPPR plates will be arranged in layers of 24 plates (6 plates by 4 plates), resulting in approximately a 12 inch by 12 inch footprint. Multiple layers will be stacked together with varying thicknesses of interspersed polyethylene placed between the layers to fine tune the neutron spectrum of the assembly. The other five experiments (Experiments 6-10) will be similar to the five baseline experiments, except that a tantalum layer will be placed next to each plutonium plate layer, thus diluting the stack and allowing for tests of the neutron cross section for tantalum.

1. INTRODUCTION

The need for epithermal and intermediate energy range critical benchmarks is an established international criticality safety data need. The U.S. Nuclear Criticality Safety Program (NCSP), a Department of Energy Headquarters Program under NA-50, convened a meeting in July of 2011 to discuss the data and experimental needs of criticality safety practitioners. The meeting was held at Sandia National Laboratories (SNL) in Albuquerque, NM with attendees from the U.S., the United Kingdom, and France. The meeting was convened under the name "Thermal Epithermal eXperiments (TEX) Feasibility Meeting," but the focus of the meeting quickly shifted more generally to the lack of experimental data in the intermediate energy range. At the TEX meeting, representatives from the U.S., U.K., and France discussed their high priority needs for benchmark data. The consensus prioritization of data needs was published in the TEX meeting minutes, as follows: ^{239}Pu, ^{240}Pu, ^{238}U, ^{235}U, temperature variations, water density variations, steel, lead (reflection), hafnium, tantalum, tungsten, nickel, molybdenum, chromium, manganese, copper, vanadium, titanium, and concrete (focusing on characterization, water content, and use as a reflector)[1].

The goal of the TEX Project is to address these recognized nuclear data needs by executing experiments with NCSP fissile assets that can be used to create critical assemblies that span a wide range of fission energy spectrums, from thermal (below 0.625 eV), through the intermediate energy range (0.625 eV to 100 keV), to fast (above 100 keV). An additional goal of the TEX project is to design critical assemblies that can be easily modified to include various high priority materials (diluents) identified by the international criticality safety and nuclear data communities.

Preliminary design for the TEX experiments evaluated the existing NCSP national inventory to find fissile material candidates that could be assembled into experimental configurations that would span a broad range of energies [2]. Due to the high priority of ^{239}Pu and ^{240}Pu cross section data needs and the results of sensitivity and uncertainly calculations conducted as part of preliminary design, it was recommended the first TEX experiments use aluminum alloyed plutonium fuel plates originally fabricated for use in Argonne National Laboratory's Zero Power Physics Reactor (ZPPR). Seven diluents were studied in preliminary design, but Ta was recommended as the first diluent for the TEX experiments. The highest priority diluents from the TEX meeting were Fe (major component of steel), Hf, and Ta. Of these three, Ta showed the highest cross section sensitivity when used as a diluent in the Pu/Al ZPPR system. Additionally, Ta provided the highest contribution to total system uncertainty of any diluent studied. Finally, the NCSP owns a large number of high quality Ta plates from the ZPPR inventory in Idaho, so diluent plates would not need to be fabricated.

2.0 EXPERIMENT DESCRIPTION

2.1 Assembly Machine

One of the two universal critical assembly machines at the National Criticality Experiment Research Center (NCERC), Planet or Comet, will be used for the TEX experiments. Planet and Comet are vertical lift machines that are used to separate a critical assembly into halves. The upper half of the assembly is supported on a stationary platen and the bottom half is supported by a movable platform. The bottom platform is raised to achieve or approach criticality and is raised until it contacts the top portion of the assembly. Figure 3.1 shows a picture of the Planet machine with a previously conducted experiment [3]. Comet and Planet are designed with a similar basic structure and can accept the same experimental fixturing.

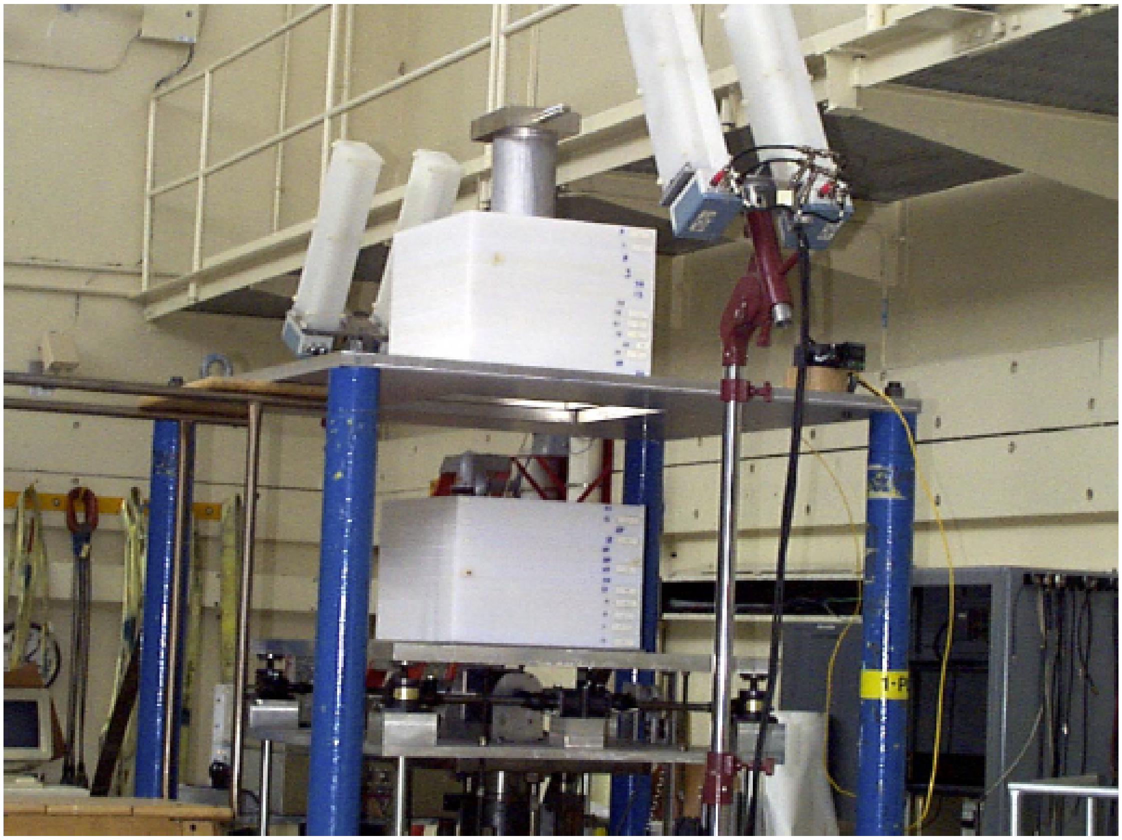

Figure 1: Planet Machine in 1998 Loaded with Polyethylene Reflected and Moderated Highly Enriched Uranium Experiment with Silicon (HEU-MET-THERM-001)

2.2 Plutonium/Aluminum ZPPR Plates

The Argonne National Laboratory procured many types of fissile material fuels for its ZPPR program. A subset of this fuel is stainless steel-clad plutonium plates that are delta-stabilized with approximately 1.1 weight percent aluminum. The Pu/Al plates of interest to the TEX program are the Plutonium Aluminum No Nickel (PANN) plates, fabricated by the Dow Chemical Company at Rocky Flats in 1960. There are three different sizes of the PANN plates, but the majority have nominal outer cladding dimensions of 1/8" (0.3175 cm) by 2" (5.08 cm) by 3" (7.62 cm). The plutonium isotopics, as presented in PU-MET-INTER-002, are 95.274 wt% ^{239}Pu, 4.537 wt% ^{240}Pu, 0.187 wt% ^{241}Pu, and 0.002 wt% ^{242}Pu [4].

2.3 Experimental Configurations

2.3.1 Plutonium Baseline Experiments

Five TEX experiments were designed to establish baseline configurations covering the thermal, intermediate, and fast fission energy regimes. For all five experiments, the ZPPR plates will be arranged in layers of 24 plates (6 plates by 4 plates), resulting in approximately a 12 inch by 12 inchfootprint. Figure 3.9 shows the nominal dimensions of a plutonium layer.

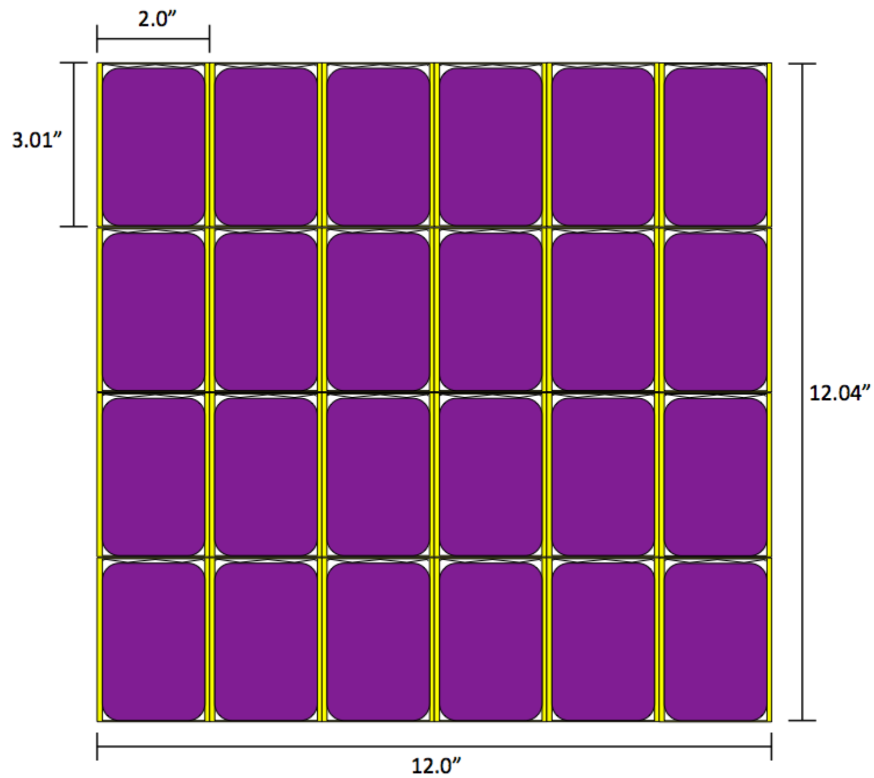

Figure 2: Single Layer of 24 Pu/Al ZPPR Plates Arranged in a Square Pattern

Multiple layers will be stacked together with varying thicknesses of interspersed polyethylene placed between the layers to tune the neutron spectrum of the assembly. The plutonium layers will be arranged on trays to facilitate stacking of the experimental assembly. There will be two kinds of trays used for the plutonium baseline experiments. The first tray, with a 0.01” aluminum bottom, is only used in the fast (unmoderated) case and has an integrated heat dispersal plate that extends 2” beyond the edge of the tray. Aluminum 6061 will be used for stability of the tray. The second tray, with a 1/16” thick high-density polyethylene (CH_2) bottom in addition to a 0.01” aluminum bottom, provides moderation, heat dispersal, and support for the Pu plates. The trays incorporate 1” of radial reflection around the edge of the plutonium layer, provided by a polyethylene frame. The trays are held together with polyethylene rivets or pins that are poly-welded in place in eight locations around the perimeter of the tray.

Figure 3 shows a cross section view of one of the five baseline configurations. The plutonium fuel meat is shown as purple and the stainless steel cladding is shown as yellow. Polyethylene is shown as blue and aluminum (Planet/Comet platform, platen, and heat dispersal plates) are shown in green.

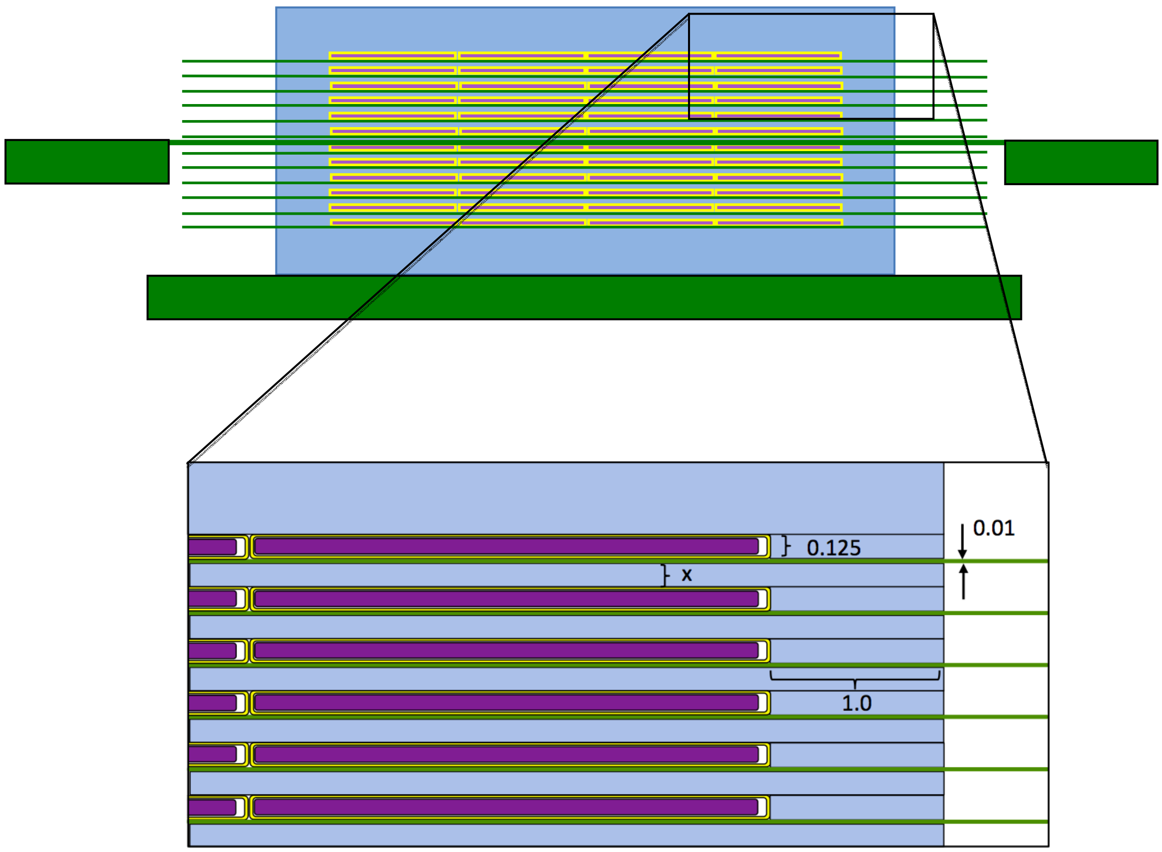

Figure 3. Experimental Configuration for Experiment 3: ZPPR Plate Layers with 0.1875 inches Interspersed Polyethylene. This experiment consists of 12 layers of Pu. The "x" in the lower figure is the variable thickness of polyethylene that can be added to the stack to tune the neutron spectrum.

2.3.2. Tantalum Diluted Critical Configurations

An additional five critical configurations will be constructed using tantalum ZPPR plates as a diluent in the five baseline critical configurations. Tantalum layers will be constructed out of 24 tantalum ZPPR plates in a similar manner to the Pu/Al ZPPR plate layers (6 plates by 4 plates). The Ta plate layers will be placed on top of and in contact with the Pu/Al layers and will be rotated 90 degrees to the Pu/Al layers to reduce neutron streaming paths.

A cross section view of the one of the Ta-diluted configurations is shown in Figure 4, with the same coloring convention as Figure 3. The tantalum plates are shown in blue. The tantalum diluted stacks are taller than the Pu baseline experiments, as the additional of the tantalum increases parasitic absorption of neutrons and the separation distance between the Pu layers, thus increasing the critical mass.

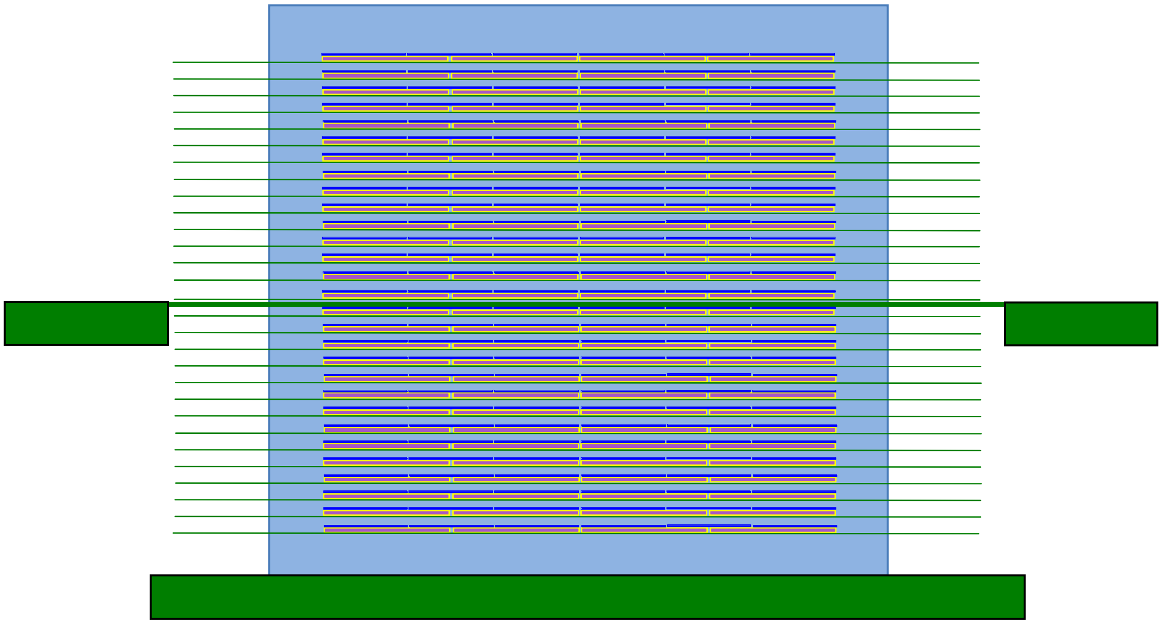

Figure 4. Experimental Configuration for Experiment 8: ZPPR Plate Layers with Tantalum and 0.1875 inches Interspersed Polyethylene. This experiment consists of 29 layers of Pu, an increase of 17 layers over the baseline case.

3.0 CALCULATIONAL MODELS OF THE EXPERIMENTS

3.1 Methodology and Code Used

The Monte Carlo neutron transport code, MCNP5, version 1.60, developed at Los Alamos National Laboratory, was used to calculate critical configurations and the corresponding neutron fission spectrum for the TEX configurations with ZPPR plates [5]. Continuous energy ENDF/B-VII.1 cross sections (.80c) were used in all MCNP5 calculations, save for a few minor constituents where ENDF/B-VII cross sections were unavailable. All materials were modeled using room temperature (293 K) cross sections.

ORIGEN, part of the SCALE 6.1 package [6], was used to decay the plutonium to July 2015 (closer to the potential experiment date).

3.2 MCNP5 Calculation Results

Iterative MCNP calculations were performed to determine critical configurations for a range of polyethylene thicknesses between fuel plate layers. Two sets of critical configurations were modeled: the first set consists of the baseline ZPPR experiments moderated by polyethylene, while the second set includes layers of tantalum plates adjacent to ZPPR fuel plate layers.

3.2.1 Baseline Pu ZPPR Experiments Results

The first set of calculations modeled the baseline case of ZPPR Pu/Al plate layers moderated by varying polyethylene thicknesses. The polyethylene thicknesses modeled were 0, 1/16", 3/16", 7/16", and 1". Table I summarizes critical dimensions for the assemblies with varying polyethylene thicknesses. The number of Pu layers reported in the table is the integer number of ZPPR plate layers required for criticality (actually made up of 24 individual ZPPR plates). The stack lengths and widths (Table I, sixth column) do not include the aluminum heat dispersal plates, which extend an additional 2" (5.08 cm) on all sides of the assembly. Calculations showed that the location of the heat dispersal plate (either on top or below the plutonium layers) caused negligible changes to k_{eff}. The aluminum heat dispersal plates are placed below the plutonium layers to assist in heat transfer.

Table I: Critical Dimensions for Pu ZPPR Plates without Ta Moderated by Polyethylene.

Experiment Number	Thickness of PE Plates (in)	Critical Mass (kg ^{239}Pu)	Number of Pu Layers	Number of ZPPR Plates	Stack Length x Width (cm)	Stack Height (cm)
1	0 (no PE)	49.8	21	504	35.59 x 35.10	12.5
2	1/16	40.3	17	408	35.59 x 35.10	13.5
3	3/16	28.5	12	288	35.59 x 35.10	12.0
4	7/16	19.0	8	192	35.59 x 35.10	15.9
5	1	14.2	6	144	35.59 x 35.10	20.5

For each critical configuration in Table I, the fission fraction as a function of energy was extracted from the MCNP output. Table II reports the Energy of Average Lethargy of Fission (EALF) and total fraction of fissions occurring in the thermal, intermediate, and fast regimes for each case.

Table II: MCNP Spectra Results for Pu ZPPR Plates without Ta Moderated by Polyethylene.

Experiment Number	Thickness of PE Plates (in)	EALF (eV)	Thermal Fission Fraction (<0.625 eV)	Intermediate Fission Fraction (0.625 eV-100 KeV)	Fast Fission Fraction (>100 KeV)
1	0 (no PE)	7.28E+04	0.09	0.17	0.74
2	1/16	5.38E+03	0.14	0.38	0.49
3	3/16	2.44E+02	0.27	0.43	0.30
4	7/16	1.57E+01	0.48	0.33	0.19
5	1	2.10	0.67	0.21	0.12

Figure 5 is a Cumulative Distribution Function (CDF) graph that visually displays the fission fraction as a function of neutron energy for the five experiments. The lower curves on the CDF graph are the less moderated systems.

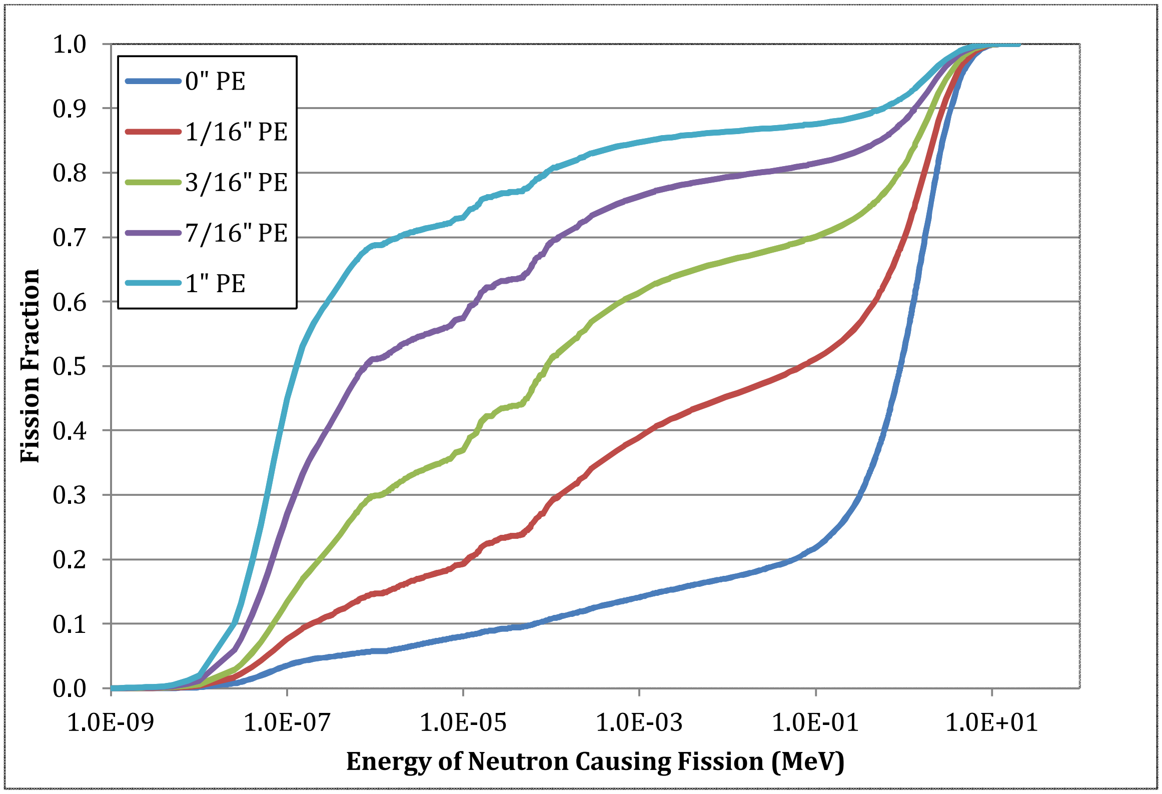

Figure 5: Cumulative Distribution Function of Fission Fraction as a Function of Neutron Energy for Pu ZPPR Plates without Ta Moderated by Varying Thicknesses of Polyethylene.

The MCNP results reported in Table II and Figure 5 demonstrate that polyethylene can be used with the Pu ZPPR plates to tune the neutron spectrum from hard (74% fast fission with no polyethylene moderator) to more intermediate or even thermal. The case with the highest percentage of fissions in the intermediate range used 3/16 in of PE, with 43% of the fissions resulting from intermediate energy neutrons.

Since one of the original TEX goals was to create intermediate spectrum experiments (where 50% or more of the fissions occur from the energy range (0.65 eV to 100 keV), additional calculations were completed that replaced the polyethylene between ZPPR layers with borated polyethylene. These scoping calculation indicated that with 3/16" borated polyethylene, a system where 52% of the fissions occur in the intermediate energy range could be created with the Pu/Al ZPPR plates, if desired for future experiments.

3.2.2 Pu ZPPR Experiments Diluted with Ta

The second set of calculations started with the baseline cases of Pu/AlZPPR plate layers moderated with polyethylene and added a layer of tantalum ZPPR plates (1/16" thick) above every Pu layer. Thesame polyethylene thicknesses from the baseline cases were modeled for the tantalum diluted cases. Table III summarizes critical dimensions for varying polyethylene thicknesses. With Ta layers, the number of Pu layers increased by 6 to 17 layers, depending on the spectrum of critical configuration.

Table III: Critical Dimensions for Pu ZPPR Plates with Ta Moderated by Polyethylene.

Experiment Number	Thickness of PE Plates	Critical Mass (kg ^{239}Pu)	Number of Pu Layers	Number of ZPPR Plates	Stack Length x Width (cm)	Stack Height (cm)
6	0 (no PE)	61.7	26	624	35.59 x 35.10	13.0
7	1/16	71.2	30	720	35.59 x 35.10	19.6
8	3/16	68.8	29	696	35.59 x 35.10	29.3
9	7/16	42.7	18	432	35.59 x 35.10	33.1
10	1	28.5	12	288	35.59 x 35.10	36.3

For each critical configuration in Table 4-3, the fission fraction as a function of energy was extracted from the MCNP output. Table IV reports the Energy of Average Lethargy of Fission (EALF) and total fraction of fissions occurring in the thermal, intermediate, and fast regimes for each case. Figure 6 is a Cumulative Distribution Function (CDF) graph that visually displays the fission fraction as a function of neutron energy for the five experiments.

Table IV: MCNP Spectra Results for Pu ZPPR Plates with Ta Moderated by Polyethylene.

Experiment Number	Thickness of PE Plates	EALF (eV)	Thermal Fission Fraction (<0.625 eV)	Intermediate Fission Fraction (0.625 eV-100 KeV)	Fast Fission Fraction (>100 KeV)
6	0 (no PE)	1.13E+05	0.07	0.14	0.79
7	1/16	1.19E+04	0.8	0.36	0.56
8	3/16	9.14E+02	0.19	0.45	0.36
9	7/16	3.38E+01	0.43	0.36	0.21
10	1	2.96	0.64	0.22	0.14

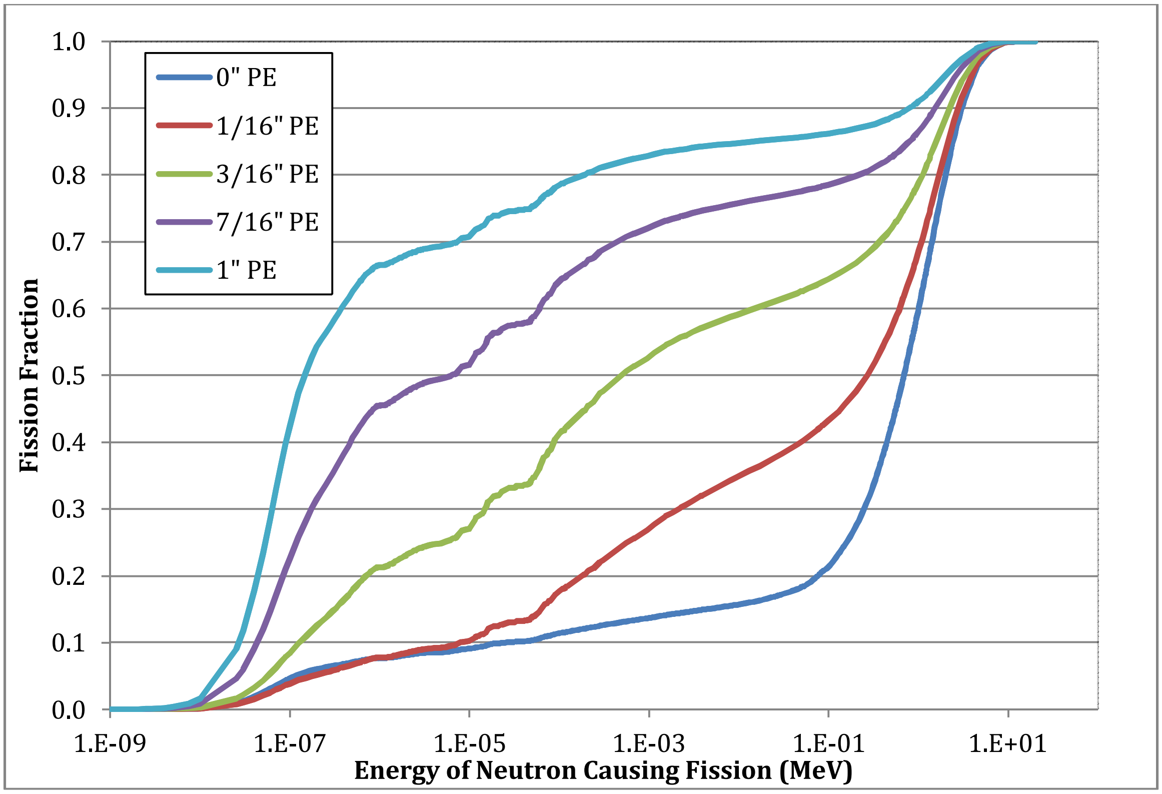

Figure 6: Cumulative Distribution Function of Fission Fraction as a Function of Neutron Energy for Pu ZPPR Plates with Ta Moderated by Varying Thicknesses of Polyethylene.

3.2.3 Preliminary Uncertainty and Bias Characterization

Sensitivity calculations were performed to preliminarily quantify the effects of various uncertainties in the experimental design on the value of k_{eff}. All of the MCNP calculations used continuous-energy ENDF/B-VII.1 cross sections, employing 6,250 generations of neutrons with 200,000 histories per generation. The first 97 generations were excluded from the statistics for each case, producing 1.23 billion active histories in each calculation. The standard deviation in the calculated k_{eff} for the individual MCNP calculation was 0.00002. Experiment 2 was used for all calculations except for the effect of tantalum impurities, which used Experiment 6. The results of the uncertainty calculations are presented in Table V and the results of the bias calculations are presented in Table VI.

Table V. Summary of Uncertainties for TEX Assembly Calculations.

Source of Uncertainty	Parameter Value used	Parameter variation in Calculation	Calculated Effect (Δk_{eff})	Standard Uncertainty of Parameter	Standard Uncertainty in Δk_{eff}
Material Mass					
Pu-239	98.87 g	0.27 g	0.00008	0.27	Negligible
Pu-240	4.697 g	-0.1 g	0.00048	-0.1	0.00048
Al	1.1584 g	-0.0464	0.00016	-0.0464	0.00016
Stainless Steel Sleeve and Plugs	24.765 g	0.046 g	0.00011	0.046	0.00011
Carbon Steel Spring	7.82 g/cm^3	0.1 g/cm^3	0.00005	0.1	Negligible
Polyethylene Plate	0.967 g/cm^3	0.005 g/cm^3	0.00086	0.005	0.00086
Polyethylene Reflector	0.967 g/cm^3	0.005 g/cm^3	0.00089	0.005	0.00089
Aluminum Mass	2.7 g/cm^3	0.027 g/cm^3	0.00026	0.027	0.00026
Geometry Dimensions					
Polyethylene Moderator Plates	13.8” × 14.0” × 1/16”	0.1” (W, L), 0.001” (T)	-0.0022	0.1”/√3, 0.001”/√3	-0.0013
Polyethylene Reflector	1” thick	0.1” (W,L), 0.001” (T)	-0.0022	0.1/√3, 0.001”/√3	-0.0013
Aluminum Plate	0.01” thick	0.005”	0.0001	0.005/√3	Negligible
Gaps between Fuel Plates	1.97” × 3.00” × 0.1205”	0.01” (W, L) 0.0005”(H)	-0.0031	0.01”/√3, 0.0005” /√3	-0.0018
Total Uncertainty	Quadrature Sum: 0.0026				

Table VI. Summary of Bias for TEX Assembly Calculations.

Source of Bias	**Parameter Value used**	**Parameter variation in Calculation**	**Calculated Effect (Δk_{eff})**	**Standard Uncertainty of Parameter**	**Standard Bias in Δk_{eff}**
Fuel Impurities	No Impurities	With Impurities	0.00024	With Impurities	0.00024
Ta impurities	No Impurities	With Impurities	-0.00006	With Impurities	Negligible
Temperature	293 ºK	15 degrees	-0.00016	15 degrees	-0.00016
Room Return	No Room	With Room	0.00017	With Room	0.00017
Total Uncertainty	Sum: 0.00057				

The sources of uncertainty from mass uncertainties pertaining to the ZPPR plate (plutonium, aluminum, steel) are very low, especially for the ^{239}Pu content. This is not a surprising result, as the plates were procured with strict ^{239}Pu requirements for use in critical experiments at ANL. A larger contributor to the delta k_{eff} was uncertainty in the polyethylene mass and dimensions. As the plates have yet to be fabricated, these perturbations were educated guesses and thus can be lessened through procurement specifications and piece-by-piece measurements. A concerted effort can also be made to lessen any gaps between plates in the assembly, which also have a relatively large effect on k_{eff}.

Fuel and tantalum impurities, temperature, and room return were shown introduce a slight positive bias to the calculations. The fuel impurity bias is an overestimate, as it assumes the worst case with the maximum impurity level from all plates and that the impurities are all carbon. The temperature is also likely overestimated, as the aluminum heat dispersal plates will largely mitigate the temperature increases across the assembly.

4.0 CONCLUSIONS

A total of ten critical configurations were designed as part of TEX-Pu final design. Five TEX experiments (Experiments 1-5) were designed to establish baseline configurations with the Pu ZPPR plates moderated by varying levels of polyethylene to cover the thermal, intermediate, and fast fission energy regimes. While MCNP5 results confirm that configurations where over 50% of the fissions occur in the fast and thermal regions can be created with polyethylene moderated ZPPR plates, the highest fraction of intermediate fissions attained was 43%. However, scoping calculations indicated that with 3/16" borated polyethylene, a system where 52% of the fissions occur in the intermediate energy range could be created with the Pu/Al ZPPR plates, if desired for future experiments.

Additionally, five experiments (Experiments 6-10) were designed that modify the baseline experiments by placing a tantalum layer next to each plutonium plate layer, thus diluting the stack and allowing for tests of the neutron cross section for tantalum. Again, MCNP5 calculations showed that critical configurations where a majority of fissions occur in both the fast and thermal energy ranges, but the highest fraction of intermediate fissions fell short at 45%, slightly increased over the baseline case. However, borated polyethylene moderator sheets can be employed to assemble a critical experiment with the ZPPR plates with greater than 50% intermediate fission fraction.

The assessment of experimental uncertainties gave very good results, especially considering the number of parts present in each configuration, with the total predicted uncertainty for the experiments as 0.0026 Δk_{eff}. Due to the discovery of a large amount of historical information regarding the Pu/Al ZPPR plates at the Idaho National Laboratory Library Archives, the experimental uncertainties attributable to the ZPPR plates were very low. Uncertainties are dominated by assembly gaps and polyethylene dimensional tolerances, which will be further reduced by careful assembly and appropriate characterization measurements.

The TEX-Pu experiments are scheduled to begin at National Criticality Experiments Research Center in 2016.

ACKNOWLEDGMENTS

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REFERENCES

1. Ellis, A. *Thermal/Epithermal eXperiments (TEX) Feasibility Meeting Minutes,* Held at Sandia National Laboratories, Albuquerque, NM, (2011).
2. C. Percher and D. Heinrichs, *IER-184 CED-1 Report: LLNL Preliminary Design for Thermal/Epithermal eXperiments (TEX)*, Lawrence Livermore National Laboratory (2012).
3. R. Brewer, "HEU-MET-THERM-001, Polyethylene Reflected and Moderated Highly Enriched

Uranium System with Silicon, Revision 1" *International Handbook of Evaluated Criticality Safety Benchmark Experiments*. NEA/NSC/DOC(95)03/I. September 2009.

4. R.M. Lell, et al. "PU-MET-INTER-002, ZPR-6 Assembly 10: A Cylindrical Plutonium/Carbon/ Stainless Steel Assembly with Stainless Steel and Iron Reflectors," *International Handbook of Evaluated Criticality Safety Benchmark Experiments,* NEA/NSC/DOC(95)03/I, (2002).
5. *Monte Carlo N-Particle, MCNP5, Version 1.60,* Los Alamos National Laboratory, (2008).
6. *SCALE 6.1: Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluation for Workstations and Personal Computers*, CCC-725, Oak Ridge National Laboratory, (2011).